

Office Action Dated Sept. 8, 2004

Appl. No. 10/070,558
Atty. Docket: 0459-0702PAMENDMENTS TO THE CLAIMS

1.-34. (Cancelled).

35. (New) A method for preparation of a polymer electrolyte membrane (PEM) for fuel cells, the method comprising the following steps:

providing an acid-doped solid electrolyte;

providing a gas diffusion cathode by:

- i) providing a first hydrophobic carbon support substrate by treatment of a carbon substrate with a hydrophobic polymer solution,
- ii) providing a first supporting layer on the first support substrate by casting a slurry onto the first support substrate, said slurry comprising carbon black, and a hydrophobic polymer,
- iii) providing a first catalyst layer on the first supporting layer by casting a slurry onto the first supporting layer, said slurry comprising carbon-supported noble metal catalysts and a polymer binder, and
- iv) doping the first catalyst layer with an acid mixture comprising a volatile acid and a non-volatile acid;

providing a gas diffusion anode by essentially utilizing the steps i)-iv) above, said gas diffusion anode comprising a second hydrophobic carbon support substrate, and a second supporting layer and a second catalyst layer; and

assembling the polymer electrolyte membrane (PEM) by sandwiching the gas diffusion anode, the solid electrolyte and the gas diffusion cathode so that the first catalyst layer and the second catalyst layer both are facing the solid electrolyte.

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36. (New) The method of according to claim 35, wherein the solid electrolyte essentially consist of a polybenzimidazole or a polymer blend comprising a polybenzimidazole and a thermoplastic polymer.

37. (New) The method according to claim 35, wherein the solid electrolyte at least 98 wt.% consists of a polybenzimidazole or a polymer blend comprising a polybenzimidazole and a thermoplastic polymer.

38. (New) The method according to claim 36, wherein the solid electrolyte essentially consist of a polymer blend comprising a polybenzimidazole and a thermoplastic polymer.

39. (New) The method according to claim 36, wherein the solid electrolyte is doped with sulfuric acid or phosphoric acid, said acid having a concentration of 40-90 wt%.

40. (New) The method according to claim 39, wherein the doping level, defined as the mole percentage of the doping acid per repeat unit of the polymer, is 250-800 where the solid electrolyte essentially consist of a polybenzimidazole, and wherein the doping level is 200-750 where the solid electrolyte essentially consists of a polymer blend.

41. (New) The method according to claim 39, wherein the doping level, defined as the mole percentage of the doping acid per repeat unit of the polymer, is 250-800 where the solid electrolyte at least 98 wt.% consists of a polybenzimidazole, and wherein the doping level is 200-750 where the solid electrolyte essentially consists of a polymer blend.

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42. (New) The method according to claim 35, wherein the first supporting layer consist of 40-60 wt% carbon black and 60-40 wt% hydrophobic polymer.

43. (New) The method according to claim 35, wherein the first supporting layer is formed by tape-casting and subsequently dried and sintered.

44. (New) The method according to claim 35, wherein the catalyst layer essentially consists of 30-55 wt% catalyst powder, preferably comprising a platinum catalyst for the cathode and a platinum-ruthenium catalyst for the anode, and 70-45 wt% polymer comprising a polybenzimidazole and a thermoplastic polymer.

45. (New) The method according to claim 44, wherein the catalyst layer is formed by slurry casting and subsequently dried and sintered.

46. (New) The method according to claim 44, wherein the polymer comprises a polybenzimidazole.

47. (New) The method according to claim 44, wherein the polymer comprises a blend of a polybenzimidazole and a thermoplastic polymer.

48. (New) The method according to claim 35, wherein said acid mixture comprises 30-70 wt% phosphoric acid or sulfuric acid and 30-70 wt% trifluoroacetic acid or acetic acid.

49. (New) The method according to claim 48, wherein phosphoric acid is used in a concentration of 2-30 wt.%, and the molar ratio of the

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phosphoric acid to the polymer contained in the catalyst layer is 5-20.

50. (New) The method according to claim 35, wherein the gas diffusion anode is prepared essentially as defined for the gas diffusion cathode, said second catalyst layer preferably comprising a platinum-ruthenium alloy.

51. (New) The method according to claim 35, wherein the assembling is performed by a hot-press.

52. (New) The method according to claim 51, wherein the hot-press is at a temperature of 80-200°C and a pressure of 0.1-1.5 bar.

53. (New) The method according to claim 36, wherein the polymer blend comprises polybenzimidazoles and one or more thermoplastic resins doped with acid.

54. (New) The method according to claim 53, wherein the polybenzimidazole is poly (2,2'-m-(phenylene)-5,5'-bibenzimidazole, the solid electrolyte comprising 10-75 mol%, of the polybenzimidazole.

55. (New) The method according to claim 53, wherein said thermoplastic resin is an ionomer prepared by sulfonation of a polysulfone, with a sulfonation degree of 5-80%.

56. (New) A polymer electrolyte membrane (PEM) for use in fuel cells, said membrane comprising the following successive layers:

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- i) a first hydrophobic carbon support substrate including a hydrophobic polymer;
- ii) a first supporting layer comprising carbon black and a hydrophobic polymer;
- iii) a first catalyst layer comprising a carbon-supported noble metal catalyst and a polymer or polymer blend as binder, said first catalyst layer being doped with an acid mixture comprising a volatile acid and a non-volatile acid;
- iv) an acid-doped solid electrolyte
- v) a second catalyst layer comprising a carbon-supported noble metal catalyst and a polymer or polymer blend as binder, said second catalyst layer being doped with an acid mixture comprising a volatile acid and a non-volatile acid;
- vi) a second supporting layer comprising carbon black and a hydrophobic polymer; and
- vii) a second hydrophobic carbon support substrate including a hydrophobic polymer.

57. (New) The polymer electrolyte membrane according to claim 56, wherein the first catalyst layer and the second catalyst layer are not identical.

58. (New) The polymer electrolyte membrane according to claim 56, obtainable by the method according to claim 35.

59. (New) The polymer electrolyte membrane fuel cell comprising a polymer electrolyte membrane (PEM) according to claim 56, which comprises a first hydrophobic carbon support substrate, a first supporting layer, a first catalyst layer, an acid doped solid electrolyte, a second catalyst layer, a second supporting layer, and a second hydrophobic carbon support substrate; said first and second

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carbon support substrates being in contact with current collection plates and, respectively, with gas flow channels are both oxidant gas and fuel gas supply.

60. (New) The fuel cell according to claim 59, wherein the polymer electrolyte membrane is as defined in claim 57.

61. (New) A method for operating a polymer electrolyte membrane fuel cell capable of operating without removal of carbon monoxide from a fuel gas before said fuel gas is being fed to the fuel cell, said fuel gas comprising a constant or intermittent carbon monoxide content of at least 0.5 vol%, said method comprising the steps of:

- i) providing the fuel cell comprising a gas diffusion cathode for reducing an oxygen-containing oxidant gas, a gas diffusion anode for oxidizing a hydrogen-rich fuel gas, and a solid electrolyte, said solid electrolyte consisting of an acid-doped membrane comprising polybenzimidazole, wherein the acid comprising a mixture of a volatile acid and a non-volatile acid,
- ii) feeding an oxidant gas to the cathode of the fuel cell, and
- iii) feeding a fuel gas, preferably a hydrogen-rich gas, to the anode of the fuel cell,

wherein the temperature of the fuel cell being 25-250°C.

62. (New) The method according to claim 61, wherein the carbon monoxide content of the fuel gas constantly or intermittently is at least 1.0 vol%.

63. (New) The method according to claim 61, wherein the oxygen-containing gas is not humidified.

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64. (New) The method according to claim 61, wherein the hydrogen-rich gas is humidified at room temperature.

65. (New) A solid electrolyte for polymer electrolyte membrane fuel cells, said solid electrolyte comprising a blend of a polybenzimidazole and one or more other thermoplastic resins doped with acid.

66. (New) The electrolyte according to claim 65, wherein the polybenzimidazole is poly(2,2'-m-(phenylene)-5,5'-bibenzimidazole (PBI).

67. (New) The electrolyte according to claim 65, wherein the electrolyte comprises 10-75 mol%, of the polybenzimidazole.

68. (New) The electrolyte according to claim 65, wherein the thermoplastic polymer is selected from polyimides (PI), polyamides (PA), polyamideimide (PAI), polyetherimides (PEI), polyarylate (PAr), poly(4-vinyl pyridine) (PVPy), and sulfonated polysulfones (SPSF).

69. (New) The electrolyte according to claim 68, wherein said thermoplastic polymer is an ionomer prepared by sulfonation of a polysulfone with a sulfonation degree of 5-80%.

70. (New) The electrolyte according to claim 65, wherein the acid with which the electrolyte is doped is an acid selected from phosphoric acid or sulfuric acid, said acid having a concentration of 40-90 wt%.